Experimental study of the mild combustion
of liquid hydrocarbons

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Abstract

A strong exhaust gas recirculation combined with air preheating generates relatively low maximum flame temperatures allowing for a better control of the furnace temperature and a lower pollutant production. While the sustainability of this combustion technology (named mild or flameless combustion) for different gaseous fuels has been extensively studied, much less information are available for liquid fuels. Consequently, this work has focused on the investigation of the sustainability of mild combustion for liquid hydrocarbons using a dual-nozzle laboratory-scale burner. Pure liquid n-octane has been used as a reference fuel, while mixtures involving also other hydrocarbons commonly found in practical fuels have been investigated to start collecting information on surrogate fuels. It has been found that the dual-nozzle configuration allows sustaining mild combustion conditions by directly injecting different liquid hydrocarbons in a mild combustion environment previously attained using a gaseous fuel. Similar results for gaseous alkanes (namely, methane and LPG) have been obtained, while using liquid hydrocarbons (namely, n-octane, n-octane/i-octane and n-octane/i-octane/n-decane) have led to somewhat different results, even if similar to each other. In particular, the mild combustion region in the T_{avg}–K_V space enlarges when using liquid hydrocarbons with respect to gaseous ones. However, in this region very low amounts of NO_x, CO, as well as negligible PAH and soot precursors have been found, therefore supporting the idea that a dual-nozzle mild combustion burner can create a suitable environment for NO_x, PAH and soot depression for a wide range of liquid wastes and low-BTU liquid fuels.
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1. Introduction

In the last decades, significant efforts have been made to achieve high thermal efficiencies in combustion processes without the adverse high NO_x emissions caused by high temperature flames [1]. It was found that a strong exhaust gas recircula-tion combined with air preheating generates relatively low maximum flame temperatures since, if the combustion air is diluted with a large amount of recirculated exhaust, the oxygen concentration in the reaction zone is strongly reduced with respect to the case of conventional burner-stabilized combustion allowing for a better control of the furnace temperature. This combustion technology is commonly defined as mild combustion [2–6] or flameless oxidation [7].

The essential principles of mild combustion are both a high preheating of the combustion air and a massive recycle of burnt gases within the furnace. The latter can be done by means of separated or

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coflowing high-velocity jets for the injection of fuel and combustion air into the furnace [6]. These jets create a low-pressure zone near the burner nozzles inducing a massive recycle of exhaust into the reactants prior the combustion reactions take place. The exhaust entrainment raises the temperature of the fresh reactants and increases the inert content of the fuel and air mixture; as a result, O_2 concentration in the combustion air quickly decreases, leading to an increase of the characteristic reaction time that becomes comparable with the characteristic mixing time, which, on the contrary, is lowered by the high turbulence generated by the high-velocity reactants jets.

This allows disappearing the flame front, that means the elimination of the temperature peak always present in conventional diffusion flames, therefore producing a fuel oxidation spread in the whole volume of the furnace and consequently a flattening of the temperature profiles inside the furnace. In other words, mild combustion occurs in a diffuse reaction zone; as a consequence, the maximum temperature rise results everywhere minimal allowing for a significant reduction of NO_x emissions produced via the thermal mechanism. Moreover, a lower maximum temperature than that found in traditional burner offers the opportunity to strongly preheat the combustion air without increasing the NO_x emissions, therefore increasing the global furnace yield.

The sustainability of mild combustion for different gaseous fuels such as methane or ethane [3,7,8], as well as mixtures of gaseous hydrocarbons and hydrogen [4,5,9], has been extensively studied; on the other hand, much less information are available concerning the sustainability of mild combustion for liquids [10,11].

Consequently, this work has been focused on the investigation of the sustainability of mild combustion for liquid hydrocarbons using a dual-nozzle laboratory-scale burner. Pure liquid *n*-octane, with its simple structure, a relatively low molecular weight and well-known properties has been used as a reference fuel. However, since a real liquid fuel is constituted by a complex mixture of hydrocarbons, surrogate fuels are commonly used both for the design of more reproducible experimental tests and for well-controlled fundamental and kinetic studies [12]. Along this line, mixtures involving also other hydrocarbons commonly found in practical fuels (namely, *i*-octane and n-decane) have been investigated.

2. Experimental section

2.1. Experimental equipment

The burner geometry (in particular the nozzles configuration for fuel and air injection) plays a key-role in mild combustion furnaces since it allows to obtain turbulence intensity and exhaust recirculation large enough to attain mild combustion conditions. In this work, a laboratory-scale burner equipped with a single high-velocity jet nozzle previously developed for mild combustion of gaseous fuels has been suitably modified and used. The original burner characteristics are discussed in detail elsewhere [3–5,8]; therefore, they are only briefly summarized in the following.

Apart from the burner, the experimental apparatus also involves an air/fuel flow-rate control section and a sampling and measurement section for temperature and exhaust compositions measurement. The laboratory-scale atmospheric burner is a vertical quartz tube made by two sections: the air preheating region and the combustion chamber (height = 350 mm, ID = 50 mm); through the closed top of the burner, thermocouples and the gas sampling line enter the combustion chamber. The combustion air is preheated by an electric oven up to 1600 K, while a separated electric oven reduces heat losses from the combustion chamber by keeping its wall temperature no more than about 150 K below the average temperature continuously detected in the combustion chamber by three type B thermocouples (named TC1, TC2 and TC3 in the following). Thermocouples TC1 and TC2 move along the combustion chamber 14 mm apart from the combustion chamber axis, while TC3 moves along the combustion chamber axis.

Air and gaseous fuel are fed through the bottom of the combustion chamber, as shown in Fig. 1a, using a single-nozzle (SN) configuration (ID = 3 mm). Since both fuel and primary air (together with diluting nitrogen when required, inlet A1 + N_2 in Fig. 1a) enter the same nozzle, a partial premix of air and fuel arises before they enter the combustion chamber. However, no fuel oxidation occurs inside the nozzle due to the short residence time (at 1000 K, it is in the range 10^{-3} to 10^{-4} s, depending on the flow rates). Figure 1a also shows the secondary air inlet (labeled A2 in Fig. 1a) that is used both for firing the burner as well as for changing the internal exhaust recycle as discussed in the following.

This SN configuration is not suitable for liquid fuels since pyrolysis would arise plugging the nozzle in spite of the small residence time; moreover, firing the burner using a liquid fuel is quite cumbersome. This required changes both in the apparatus configuration and in the firing procedure, as discussed in the following.

The apparatus has been modified by implementing the double-nozzle (DN) inlet configuration shown in Fig. 1c, where the preheated air enters the combustion chamber through the bottom nozzle, while the liquid fuel is injected as a well-dispersed and homogeneous spray through a lateral water-cooled plain jet airblast atomizer
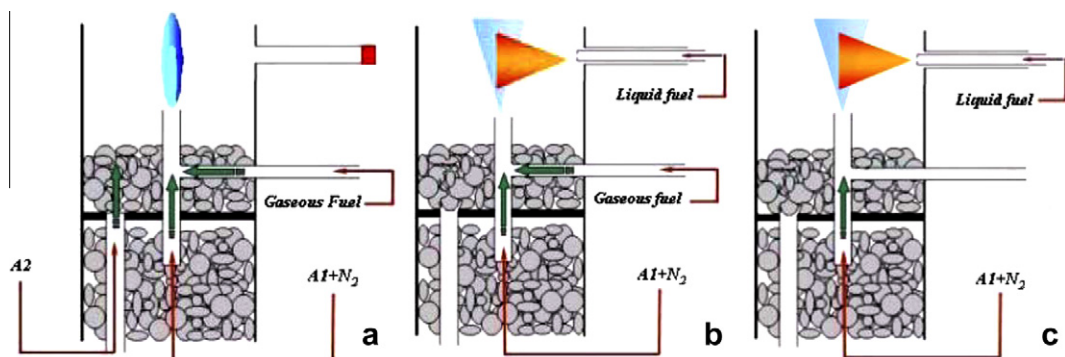
Fig. 1. Reactants feeding system: (a) SN gas-fuel feed; (b) DN liquid- and gas-fuels feed; (c) DN liquid-fuel feed. A1: primary air; A2: secondary air.

(fuel feeding pipe $ID = 0.4$ mm). The two jets interact perpendicularly and mix to each other in a high turbulence region.

Since firing the laboratory-scale burner in the DN configuration with liquid fuels is not easily done in practice, the burner has been always fired with a gaseous fuel in the SN configuration (Fig. 1a). Once mild conditions have been attained in the SN configuration, the transition from the SN gas-fuel configuration (Fig. 1a) to the DN liquid-fuel one (Fig. 1c) has been carried out by reducing progressively the gas flow rate fed to the bottom nozzle while increasing the liquid flow rate from the atomizer (Fig. 1b); for this reason, during the transition from a SN to a DN configuration two different fuels have been fed contemporarily to the burner, a gas from the bottom nozzle and a liquid from the lateral injection point, reducing step-by-step the gas to liquid ratio.

Exhaust have been sampled, dried and analyzed with two on-line instruments, namely a gas analyzer Horiba PG-250 (for NO_x, O_2, CO and CO_2 measurement) and a GC-FID Perkin Elmer Clarus 500 (for C1–C6 unburned hydrocarbons measurement); maximum deviations of about 1 ppm and 2 ppm from the mean value have been estimated for NO_x and CO concentrations, respectively. C1–C6 hydrocarbons concentration can give a rough indication of highly toxic pollutants production since they are PAH and soot precursors [2].

2.2. Mild combustion key parameters

This experimental apparatus allows for investigating the influence of two of the main operating parameters for mild conditions achievement, namely: average combustion chamber temperature (T_{avg}) and dilution ratio (K_V), defined as the ratio between the recycled exhaust and the incoming air and fuel flow rates [3,7,9].

While in real-size burners with internal exhaust recycle the K_V value is defined by the burner geometry and the air and fuel jets flow-rates, in the laboratory-scale burner the K_V value can be

changed by feeding either a secondary air stream around the bottom jet (which would reduce the exhaust entrainment and consequently the K_V value) or some inert gas together with the combustion air (which would mimic a further exhaust entrainment and consequently increase the K_V value).

Several calculations previously performed with a general-purpose code for computational fluid dynamics [3,9] showed that the maximum K_V value arising from the internal exhaust recycle without any secondary air or inert gas addition (R) is equal to about five for all the conditions investigated in this work; this value is reached at about 5.5 cm from the bottom nozzle tip, where the lateral spray-jet has been located.

It is possible to estimate the maximum K_V value in the laboratory-scale burner when either an external recycle or a secondary air inlet is present as:

$$K_V = \frac{R - S/A}{1 + S/A} + \frac{(I/A) \times (1 + R)}{(1 + F/A) \times (1 + S/A)} \qquad (1)$$

where A is the flow rate of the primary air, S of the secondary air, I of the inert gas, and F of the fuel. As expected, when neither secondary air nor inert gas are fed to the laboratory-scale burner, the previous relation leads to $K_V = R$.

The practical identification of mild conditions boundaries requires the definition of some threshold values either for pollutant emissions or for temperature gradients in the combustion chamber. Following previous studies [4,5], clean mild conditions have been defined as characterized by $NO_x < 30$ ppm and $CO < 50$ ppm in the exhaust.

3. Results and discussion

3.1. SN vs. DN configuration

At first the influence of the new DN configuration and that of the new firing procedure have been

investigated using methane as a reference fuel for the SN configuration. Several experiments have been carried out performing the transition from SN to DN configuration by injecting from the lateral nozzle methane (to investigate the influence of switching the fuel feed from the bottom to the lateral nozzle), LPG (to investigate the influence of changing the fuel properties from methane to heavier alkanes, which perform quite similarly in the high temperature combustion region, without involving droplet evaporation), or *n*-octane (which is a reference fuel commonly used for building surrogate fuels [12]). For the investigated fuels, depending on the experimental conditions to be realized, maximum flow rates of about 2.5×10^{-4}, 1.6×10^{-4} and 7.3×10^{-5} mol/s have been used for CH$_4$, LPG and *n*-octane, respectively; the air flow rate has been evaluated according to both the reaction stoichiometry and the required air excess.

In all the experiments the firing procedure summarized in Fig. 1a–c has been used apart from nitrogen injection from the lateral nozzle, which is required only for liquid fuels atomization; in this case the atomizer has been cooled at 333 K and flushed with 2 Nl/min of nitrogen to generate a spray of small and homogeneous fuel droplets with a short penetration distance. Concerning the liquid droplets, an SMD of about 35 μm has been estimated for the fuels here investigated. Due to the experimental apparatus characteristics, the thermal power values investigated changed with the fuel from about 0.2 kW for CH$_4$, to about 0.3 kW for LPG, up to about 0.4 kW for liquid fuels.

Figure 2 shows a comparison among SN to DN transitions for the three different fuels. When CH$_4$ is used, stable clean mild conditions can be achieved without any significant variation in both temperatures and NO$_x$ emissions. Stable clean mild conditions can be also achieved using LPG, even if in this case temperatures and NO$_x$ progressively rise when the LPG/CH$_4$ ratio increases due to the increase of the thermal power input. A sim-

ilar behavior has been found when using *n*-octane, even if in this case the temperature increase is partly limited by the cold nitrogen used to spray the fuel as well as by the fuel droplets evaporation; in this run, the progressive switch from the CH$_4$/*n*-octane = 3/1 (75:25% by vol.) condition to the pure liquid-fuel feed has been done in order to increase the thermal input only of about 25%, resulting in a slight increase in the average furnace temperature. However, in spite of the average temperature in the combustion chamber is almost the same for LPG and *n*-octane, the latter shows a higher NO$_x$ emission; even if local hot spots can be created by spray combustion hot spots and can contribute to increase the NO$_x$ emissions, the measured trend should be mainly ascribed to the NO$_x$ formation through the Prompt mechanism [13] in the region close to the evaporating droplets, where large fuel/air ratio values are expected. While this mechanism plays a negligible role for LPG, it is important for *n*-octane, as confirmed by simulations carried out in a simple ideal reactor configuration with a detailed kinetic mechanism [14].

The results summarized in Fig. 2 clearly indicate the possibility of operating the fuel switch to DN configuration when the combustion chamber is operating in SN mild combustion conditions without creating any significant instability. This means that a SN laboratory-scale mild burner can be easily retrofitted to operate in stable mild combustion conditions with both a different nozzle layout and a direct liquid fuel injection, therefore suggesting the possibility to perform the same retrofit on real-size mild burners. This would extend their operability in terms of energy saving, fuel availability, emissions control and cost reduction.

Once assessed the reliability of the DN configuration, the influence of T_{avg} and K_V on the mild combustion attainment in this configuration has been investigated and compared (in a typical T_{avg} vs. K_v diagram) with literature data of CH$_4$ mild combustion in SN configuration [5] (to ensure a
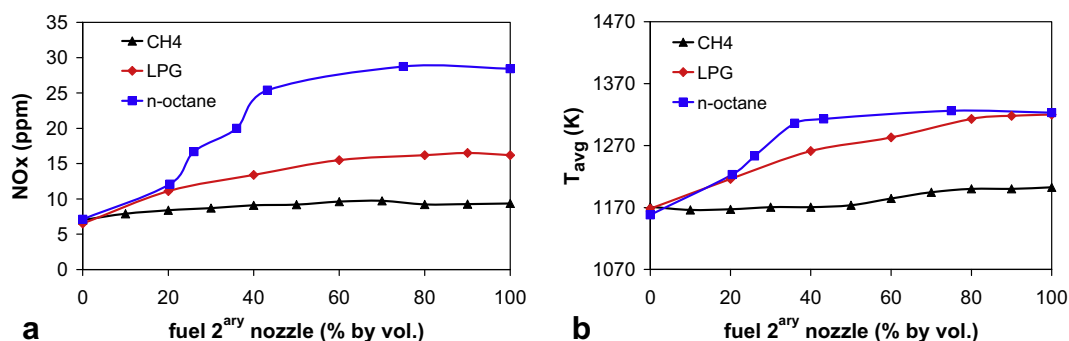

Fig. 2. NO$_x$ emissions (a) and average temperature (b) as a function of the lateral/bottom fuel ratio (0% = SN; 100% = DN). $T_{pre-heater}$ = 1273 K; excess air = 20% by vol.

direct comparison of the results, all data have been reported @ 3%O_2). The results of such a comparison are summarized in Fig. 3, where the mild combustion region for the SN configuration is bounded in the high temperature region by a zone where mild conditions can be sustained but thermal NO_x production becomes significant, while a no combustion (or extinction) zone and a flame combustion zone bound the mild combustion region for T_{avg} and low K_V values, respectively. The experimental results obtained with the DN configuration show that stable mild conditions can be sustained also outside the mild combustion region for the SN configuration, enlarging the mild operating ranges in terms of K_V, until about 3, while producing only small changes about the lower T_{avg} threshold (at about 1130 K). It should be noted that the decrease of the lower dilution threshold up to about $K_V = 3$ confirms literature experimental data of multi-nozzle burners [7]. This confirms that a SN burner configuration represents the worst case in terms of mild combustion feasibility since combustion can start before the reactants dilution is achieved [5].

It is worth noting that LPG shows practically the same threshold values as CH_4 in spite of its higher reactivity, flame speed and thermal power input. This means that the DN configuration allows for the achievement of an optimal reactants and thermal dilution before the combustion reaction starts, thus reducing thermal peaks within the combustion chamber and suggesting that the thresholds found in these experiments can be probably extended to other small gaseous alkanes and their mixtures, such as Natural Gas.

During the transition from SN to DN configuration the fuel conversion has been found to be always complete, with no unburnt hydrocarbons and negligible CO emissions.

3.2. Liquid n-octane mild combustion

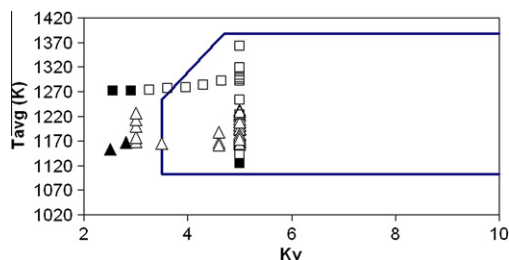Several tests have been dedicated to investigate the influence of air excess and pre-heater temper-ature ($T_{pre-heater}$) on the mild combustion sustain-ability of n-octane with reference to CO formation (because NO_x emissions, in the conditions here reported, are always of the order of 10 ppm). Figure 4 summarizes CO emissions as a function of T_{avg} for three air excess values at $T_{pre-heater} = 1223$ K, while Fig. 5 shows CO emissions vs. air excess for two K_V values at $T_{pre-heater} = 1123$ K. Data reported in Fig. 4 are obtained adjusting the thermal input of the system by means of a reduction of the reactants flow rates. We can see that both air excess and pre-heater temperature have a small influence on the CO formation; the CO threshold value is always reached when the temperature decreases significantly or when the dilution ratio increases significantly, leading to a too low oxygen concentration to ensure a complete fuel conversion (for both figures, similar conclusions can be drawn even if the data are expressed in terms of CO yield instead of CO concentration).

Figure 6 reports the influence of K_V on the mild combustion emissions, where the possibility of operating with K_V values as low as 1.5, which is significantly lower than the minimum K_V value found for CH_4 and LPG, is clearly identified. In all the
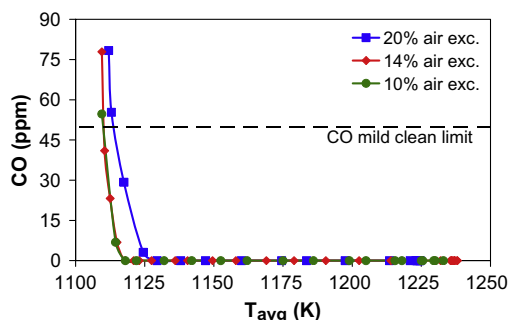

Fig. 4. CO emissions as a function of air excess and T_{avg} in DN n-octane mild combustion; $T_{pre-heater} = 1223$ K, $K_V = 7.5$.
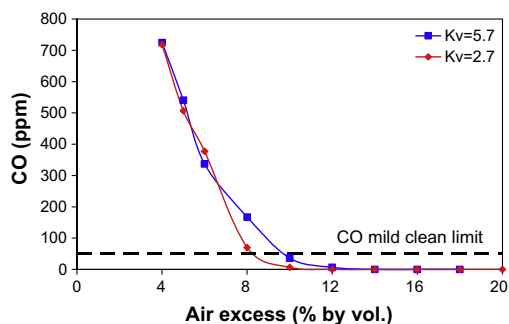

Fig. 3. Experimental results for CH_4 ($\triangle$) and LPG ($\square$) with DN configuration; solid line represents the mild combustion region boundary for CH_4 with SN config-uration [5]. Empty and full symbols represent mild combustion and non-mild conditions, respectively.

Fig. 5. CO emissions as a function of air excess and K_V in DN n-octane mild combustion; $T_{pre-heater} = 1123$ K.
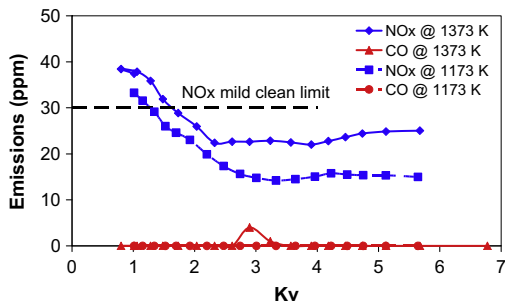
Fig. 6. Emissions trends for DN n-octane mild combustion with different $T_{\text{pre-heater}}$ (Air excess = 14% by vol).

mild combustion DN runs with n-octane, NO_x emissions equal to about 10–15 ppm have been found together with very low CO concentrations (small CO fluctuations, in the range of the experimental error, have been sometimes detected, but were always very low compared with the corresponding threshold limit value, that is 50 ppm) and practically no emissions of unburnt hydrocarbons.

The mild operating map for n-octane is shown in Fig. 7, where the empty symbols indicate clean mild combustion conditions, while full squares and full triangles represent conditions that are above the NO_x and CO threshold limits, respectively. The clean mild combustion region is bounded by a lower average temperature value of about 1110 K, a lower K_V value equal to about 1.5, and an upper average temperature value of about 1290 K. This upper value is due to the NO_x formation and it is quite lower than the corresponding threshold found for gaseous fuels (see Fig. 3). An increase of the excess air ratio, as shown in Figs. 4 and 5, can have a slight positive effect on lower boundary limits, enhancing the clean mild combustion stability at low K_V and low average temperatures values; on the other hand, a too large excess air

amount, contributing to the Thermal-NO_x formation, can slightly reduce the upper average temperature limit of the clean mild combustion region.

The difference between gaseous and liquid fuels in the upper temperature threshold cannot be explained by the presence of the spray-jet since the main structure of the flow field in both cases is established by the air jet on the bottom of the combustion chamber. However, the presence of fuel droplets that must evaporate before burning creates a region with a large fuel/air ratio where prompt-NO_x mechanism could play a significant role and explain this difference. In fact, while thermal-NO_x formation rate strongly depends on temperature and only to a less extent by oxygen concentration [4], prompt-NO_x are formed by the reaction of nitrogen with hydrocarbon radicals in the fuel rich regions at temperature below 1773 K [13]. As shown in Fig. 8, even if a qualitative relationship can be found between air pre-heater temperature and NO_x emissions underlying the role of the thermal-NO_x route, at such moderate temperature values the NO_x concentration values are too high to be explained only by the thermal-NO_x route, accordingly with the previous discussion of the results reported in Fig. 3 for LPG and n-octane mild combustion.

Almost no differences have been found between gaseous and liquid fuels for the lower average temperature threshold (see Figs. 3 and 7), since this limit is due to the CO formation and it is scarcely influenced by the fuel composition being controlled by the combustion chamber temperature and its gradients along the burner. A somewhat different consideration can be done for the lower K_V threshold that, for both gas and liquid alkanes, is determined by a gradual increase of the NO_x emissions (see Fig. 6) without any increase of the average combustion chamber temperatures. The reason is that a decrease of K_V involves a lower air/fuel dilution with the exhaust, thus leading to a local temperature increase in the lower part of the combustion chamber, as confirmed by measurements carried
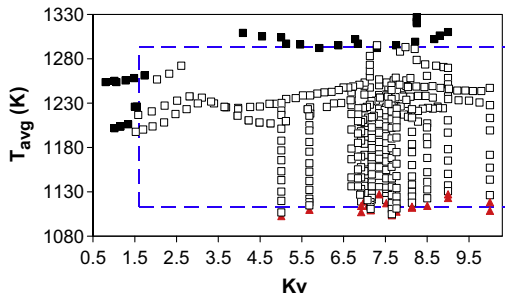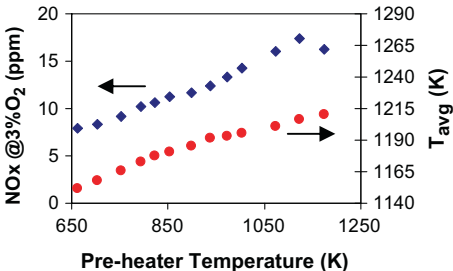

Fig. 7. Mild combustion region for n-octane in DN configuration (Air excess = 10% by vol.). Empty symbols: combustion conditions inside the mild combustion region; Full squares and full triangles represent conditions outside the NO_x and CO threshold limits, respectively.

Fig. 8. NO_x emissions and average temperature as a function of the $T_{\text{pre-heater}}$ for n-octane mild combustion; $K_V = 5.7$.
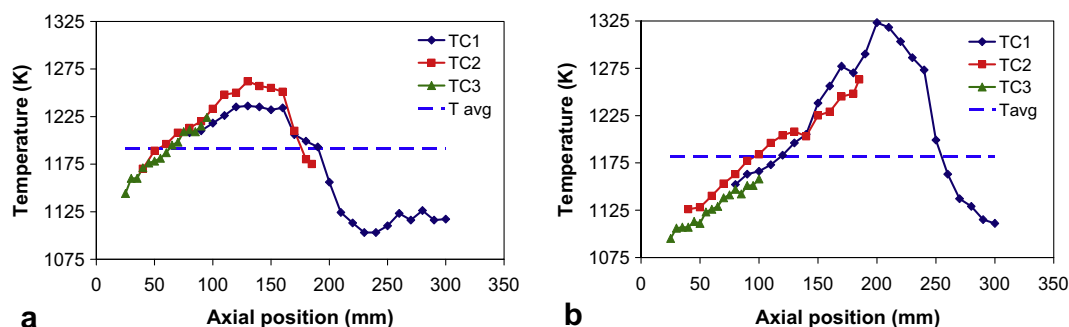
Fig. 9. Temperature profiles measured along the combustion chamber at $K_V = 5.7$ (a) and 2.7 (b) for *n*-octane mild combustion in DN configuration (lines with symbols) and average temperatures (dashed lines); $T_{pre-heater} = 1173$ K.

out along the combustion chamber. Such measurements (which are affected by an uncertainty lower than 2%) are summarized in Fig. 9 for two different K_V values (similar trends have been observed for all the investigated K_V values). We can see that, as expected, reducing the K_V value leads to larger maximum temperature values while keeping almost the same average temperature value.

However, this threshold value has been found lower for *n*-octane than for gaseous alkanes. This difference can be explained considering that the overall combustion rate of a liquid hydrocarbon can be significantly reduced by the droplets evaporation rate, therefore reducing the Damkholer number ($Da = t_m/t_c$), which is the ratio between a characteristic mixing time, (t_m) and a characteristic combustion time (t_c) [4]. Mild combustion conditions, which require the Damkholer number to be low enough, are usually attained by increasing the jet velocity (which reduces the mixing time) and lowering the overall combustion rate by reducing the oxygen concentration trough exhaust recycle (which increases the combustion time). In the case of liquid *n*-octane combustion, the overall combustion rate is reduced also by the droplet evaporation kinetics, resulting in the possibility of sustaining mild combustion conditions even with larger oxygen concentrations, that is, with lower K_V values.

This could mean that the injection of a liquid hydrocarbon in the DN configuration can sustain mild combustion of liquid wastes and/or low-BTU liquid fuels at low costs and with low pollutant emissions. In this respect, it is worth remembering that no emissions of unburnt hydrocarbons (that can be roughly considered as the precursors of PAH and soot) have been found in all the experiments.

3.3. Liquid hydrocarbons mild combustion

To verify the possibility to obtain a stable mild combustion also for liquid hydrocarbons mixtures more similar to commercial liquid fuels (following
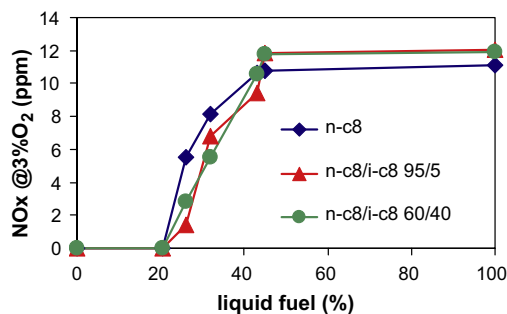

Fig. 10. NO_x emissions as a function of the lateral/bottom fuel ratio (0% = SN; 100% = DN). $T_{pre-heater} = 1173$ K.

the surrogate fuels approach [12]), various liquid mixtures have been tested in the DN configuration.

As surrogate liquid fuels commonly contain linear and branched alkanes, the first mixture tested involves *n*-octane and *i*-octane. As shown, for the sake of example, for NO_x emissions in Fig. 10, the presence of the branched isomer does
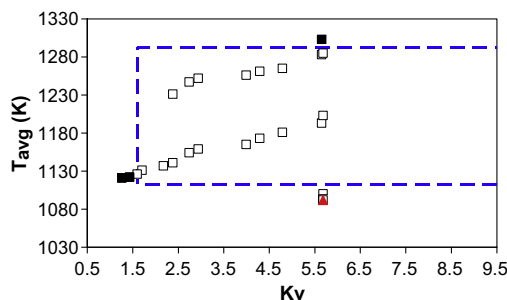

Fig. 11. Experimental results for a liquid mixture constituted by *n*-octane/*i*-octane/*n*-decane (36/24/40 by vol.) with DN configuration; dashed line: mild combustion region boundary for liquid *n*-octane. Empty symbols: combustion conditions inside the mild combustion region. Full squares and full triangles represent conditions outside NO_x and CO threshold limits, respectively.

not affect significantly mild combustion conditions with respect to the aforementioned *n*-octane results.

Moving a further step towards real fuels, a liquid mixture constituted by *n*-octane, *i*-octane and *n*-dodecane (36/24/40 by vol.) has been tested. Also this mixture shows a similar behavior, being its transition from SN to DN configuration characterized by NO_x emissions lower than 30 ppm and negligible CO emissions. As shown in Fig. 11, the results obtained using this mixture fully compare with the previously identified mild operating map of *n*-octane. This means that, as found for the mixtures constituted by octane isomers, in mild conditions the considered liquid hydrocarbons show a similar reactivity.

4. Conclusions

In this work the sustainability of mild combustion for liquid hydrocarbons has been investigated using a DN laboratory-scale burner and methane as a reference fuel for the SN configuration.

It has been shown that the DN configuration allows to sustain mild combustion conditions by directly injecting different liquid hydrocarbons in a mild combustion environment previously attained using a gaseous fuel.

DN configuration provides similar results for gaseous alkanes (namely, methane and LPG), while using liquid hydrocarbons (namely, *n*-octane, *n*-octane/*i*-octane and *n*-octane/*i*-octane/ *n*-decane) leads to somewhat different results, even if they are similar to each other. This means that the mild combustion characteristics seem more influenced by the physical state of the fuel (that is, gas or liquid) than that by the chain length of the hydrocarbon (that is, C1–C4 or C8–C10).

In particular, the mild combustion region in the T_{avg}–K_V space enlarges when using liquid hydrocarbons with respect to gaseous ones shar-

ing the same class (that is, alkanes). In this region very low amounts of NO_x, CO, as well as PAH and soot precursors are produced.

These findings support the idea that a DN mild combustion burner can create a suitable environment for NO_x, PAH and soot depression, allowing the use of a wide range of liquid wastes and low-BTU liquid fuels, even with unsteady composition or coming from different sources.

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